Correlation between Stick - Slip events and contact charging in the dynamic friction at nano-scales

G. Ananthakrishna

Materials Research Centre

Indian Institute Science

Bangalore - 560012

Collaborator

Jagadish Kumar

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MRC, IISc, Bangalore



- Introduction
- Experimental results of Putterman (PRL, 2000) unexplained, Conclusions - intriguing
 - SFA on PMMA : Correlation between stick slip events and charge transfer. Ascribe friction to bonds formed due to contact electrification.
- Modelling Specific issues. Contact mechanics, visco-elasticity, visco-plasticity, contact electrification.
- Results.
- Summary and Conclusion.

Leonardo da Vinci, a self taught genious (April 15th 1452– May 1519)

Self Portrait of Leonardo da Vinci, *1512*, First quantitative frictional studies by Leonardo da Vinci and Sketches from the Codex Atlanticus and Codex Arundel.





Sketches from the Codex Atlanticus and the Codex Arundel.

- (a) The force of friction between horizontal and inclined plane
- (b) The influence of apparent contact area upon the force of friction.
- (c) The force of friction on a horizontal plane by means of a pulley.
- (d) The friction torque on a roller an half bearing.

Anatomist

Muscles of the arm and shoulder in rotated views, c. 1510

Anatomical Studies, folio 141v



Figure 2-2: Manilo of the orna and skuilder in rotated views, c. 1510, Anatomical Similar, 550-1410

vertebral column, c. 1510,

Anatomical Studies, folio 139v

Ligare 5-1: The vertebrai volume, v 1510. Amitembiai Studies, frie 1390 Fetus within the womb,

c. 1510-12

Anatomical Studies, folio 198r



Mechanical Engineer



Figure 5-5: Rotary tall boaring. Godex Madrid 1, felio 20m model In Much Technic Manual 1404

Figure 8-3: Water powered rolling with Codex Atlanticm, filie 10:

Figure 2-31 Two-whaled kotst. Codox Atlanticus, folio 501-

Rotary ball bearing Codex Madrid I folio 20v

water-powered rolling mills **Codex Atlanticus** folio 10r

Two-wheeled hoist **Codex Atlanticus** folio 30v

Aerodynamics Engineer



Figure 5-1: Design for flying numbers, Mr. B, folio 14v

Design for flying machine

Ms, B, folio 74v



Working model of a flying machine

Museum of history of sciences, Florence

Scientist: Turbulence



Figure 6.4: Turbulent wakes hebrid a reatingular plank, z. 1569–11. Window Collision, Landscapes, Plance, and Water Studies, files 42r

Turbulent wakes behind a rectangular plank

c. 1509-11, Windsor, Collection

Architect



Figure 7-7: May of the Channa valley, 1504, Window Collection Desarrays and Mixelianeous Papers, Vol. IV July 4358

Map of the Chiana valley

1504, Windsor Collection



Figure 2-5: Design for Centralized "Temple," c. 1488, Ms. Ashburnham I, folio 5v

Design for Centralized "Tample,"

c. 1488, Ms. Ashburnham I, folio 5v

- Friction is the energy dissipated when two surface are in relative motion.
- It arises due to different fundamental causes.
 - Mechanical in nature: Interlocking of asperities at the sliding interface.
 - Chemical in nature : Adhesion between two solid in contact.
- Earlier lubricants had been used to reduce friction between sledges used for transportation.



Painting from the tomb of Tehuti-Hetep, El-Bershed (1880 BC). The colossus is secured on a sledge but there is no roller or levers.

Friction - scale dependent - macro, micro, nano scales.

Friction and Stick-Slip: Leonardo da vinci, 16th century Frictional resistance $f \propto load$, \rightarrow is independent of area S $F = \sigma_{y} S_{eff}$ Amonton 1699, Coulomb 1781 : Amonton - Coulomb laws: Minimum force required to move $f_s = \sigma_s S_{eff}$ $f_s =$ pulling force, F = normal force, S = area, μ_s = static friction coefficient. $\rightarrow f_s = \mu_s F$ M For constant velocity v, $f_d = \boldsymbol{\mu}_d F$ dynamic friction coefficient. μ_s and μ_d independent of S and F. H d 0.5 $\mu_{s} = \mu_{d} = -$ 0.4 4 0,3 з f/Mg μ_=0.24 Inil 1 0.2 2 0.1 1 0 rest loading steady sliding 0 8 12 16 20 0 Tatick Mg (N) time

Bowden Tabor (1950):

 $\sigma_s < \sigma_y$: are material property.

 $F = \sigma_y S_{eff}$ $f_s = \sigma_s S_{eff}$

Thus, $\mu_d = \frac{\sigma_s}{\sigma_y} \rightarrow \text{Amonton}$ - Coulomb law. At constant v, time of contact reduces hence,

 $\mu_d = \mu_d(v)$, decreases. as v increases.

In velocity weakening regime, any fluctuation is unstable \rightarrow Stick-Slip. Stuck state is due to elastic loading and Slip state due to stress relaxation.



(a) M = 2.1 kg, $k = 1.5X10^4$ N m⁻¹, and $V = 10 \ \mu m s^{-1}$ in paper-on-paper system. (b) Creep plot of the slider displacement x Vs t for M = 0.32 kg, $k = 1.5X10^4$ N m⁻¹, and $V = 5 \ \mu m s^{-1}$.

The crucial physical ingredient responsible for stick-slip behavior is "velocity-weakening" phenomena.



(a) Velocity-weakening law . A slight uncertainty in the stiffness leads to unstable motion i.e shown. (b) Schematic view of two rough surfaces. (c) Optical visualization of two rough epoxy resin blocks.

Friction process is complex :

adhesion, wear, interfacial layer, plastic deformation, smooth, roughness, contact electrification etc., are contribute to friction.

-Plastic deformation - contact radius , load - Hertz vs JKR, multiple scales in roughness





Electronic states deep in the gap of the polymer arising from side-groups. The location of the lowest occupied molecular orbital (LUMO) to highest occupied molecular orbital (HOMO) with respect to E_F is shown.

Surface Force Apparatus (SFA) is a useful technique to study friction at these scales. AFM, R ~ nm, F_n~ μ N-mN, σ ~ GPa;

SFA, $R \sim \mu m$ - mm, F_n = a few mN, $\sigma \sim 50 MPa$.

- When a microcantilever with a nano-scale tip is scanned laterally over a surface to measure the nano-scale force it exhibits stick-slip motion.
- The nature of stick-slip depends on probe stiffness, structure of the tip, surface energy and scan parameters (load, velocity, etc.)

Experiment:

- SFA experiment of Budakian and Putterman (Phys. Rev. Lett., 85, 1000 (2000))
 Friction ascribed to the formation of bonds arising from contact charging
- The cantilever of a SFA was dragged with a velocity of a few $\mu m/s \sim 10 \mu m/s$ on polymethylmethacrylate (PMMA) surface.
- $The R of the gold ball 0.5 mm, V a few \mu m/sec \sim 10 \ \mu m/sec.$
- Typical measured charge density ~ 10^8 charges/mm².





Image of charge during stick-slip motion

Experiment: contd...

Putterman et al.(2000): main results to be explained : Frictional sliding is due to contact charging - bonds are formed. (1) correlation between force (stick-slip events) and charge transfer.



(2) the total force is proportional to the total charge deposited over a scan length. Scale factor α .

(3) The value of $\alpha \sim 0.4 \text{ eV} / \text{\AA}$; α constant for $68 \leq F_n \leq 106 \text{ mN}$.

- α same for smooth sliding,
- fewer stick-slip events of larger magnitude for higher F_n

(4) $\alpha \sim 0.4 \text{ eV}/\text{\AA}$ is close to the energy window for transfer of charge from Fermi level

Why doubt their claims?

A) Force of Attraction due to dipole layer

 $F_c = (\mu \pi a^2 \sigma) / (2\epsilon_0 \kappa)$

Hertz radius is $a_H^3 = 3F_n R/4E^*$;

Using R = 0.5mm; $F_n = 0.1N$; $E^* = 1.3GPa$;

 $a_H = 22\mu m$

Using this and

 $\sigma = 10^8 esu/mm^2 = 1.67 * 10^{-5} C/m^2; \\ \epsilon_0 = 8.85 * 10^{-12} C^2/Nm^2; \\ \kappa = 3.5$

It turns out $F_c = 10^{-9} N$, but $\delta F \sim mN$

B) Stress
$$\tau = F_n / (\pi (2.2)^2 * 10^{-10}) = 6.9 * 10^7$$

 τ_y for PMMA is 10MPa

 τ_y for Gold is 80MPa Thus asperities are plastically deformed.

Toy Model

Our aim is to build a simple model to explain the major features of the experiments.

A toy for stick-slip and couple to charging and charge transfer equation.

$$\ddot{X} = F - A\dot{X} - \frac{\dot{X}/v_m}{1 + (\dot{X}/v_m)^2} - f_{ch}\Sigma^2$$
(1)

$$\dot{\Sigma} = \frac{1-\Sigma}{\tau} - \frac{\dot{X}\Sigma}{a}$$
(2)

$$\dot{\Sigma}_d = \frac{\dot{X}\Sigma}{a} \tag{3}$$

$$\dot{F} = v_a - \dot{X} \tag{4}$$

- Equation for dimensionless displacement X of the gold tip.
- $A\dot{X}$ dissipative term, III term the velocity weakening law that has maximum at v_m .
- The last term is frictional resistive force from electrostatic adhesion

The parameters used in the results reported here are $v_a = 0.02$, $v_m = 0.01$, $\tau = 2.0$, a = 1.57 and $f_{ch} = 2.56 \times 10^{-7}$.



Figure 3: (a,b) Plots of force verses displacement for A = 3.0 and 7.0 respectively, and (c, d) displacement and cumulative charge as a function of time corresponding to (a).

Results: Contd...



Figure 3: (e,f) Plots of X verses time and Σ verses time for A = 3.0.

- Results clearly displays the correlation between the displacement and the cumulative charge deposited on the PMMA surface and both has same slope.
- Fewer stick slip events for larger F_n
- The established correlation between X and Σ_d also suggests that the authors interpretation that macroscopic friction arises from 'collective effects of bonds' formed due to charge transfer is not entirely warranted.

Full Model

- Include all relevant physical mechanisms:
 - (a) Singe asperity
 - (b) JKR vs Hertz
 - (c) Visco-elasticity
 - (d) Plastic deformation
 - (e) Adhesion due to contact charging





Full Model

(a) Hertz relation for the area of contact - contact radius and depth

$$A_n = \pi a^2 = \pi [3RF_n/4E^*]^{2/3} = \pi Rz \tag{5}$$

For $E^* = 2GPa$; $F_n = 0.1N$; R = 0.5mm $a_H \sim 2.67 * 10^{-5}m$ (b) JKR radius -

$$a^{3} = \frac{3R}{4E^{*}} \left(F + 3\pi\gamma R + \sqrt{6\pi R\gamma F + (3\pi R\gamma)^{2}} \right)$$
(6)

 $a_{JKR} \sim 3.4 * 10^{-5}$ for $\gamma = 0.1 J/m^2$ and since $a_{JKR} \simeq a_H$, use a_H

- (c) Visco-elastic effect $G \longrightarrow G + \eta \dot{\epsilon}$
- (d) Plasticity of interface asperities: $\dot{\epsilon}_{ve} \longrightarrow \dot{\epsilon}_{ve} + \dot{\epsilon}_{pl}$.

(e) Contact charging contribution - Using this contact area of charging, the attractive force is given by $\pi R z \sigma^2 / 2\epsilon_0 \kappa$.

(f) For contact radius ($a \sim 28 \mu m$ for $F_n = 0.1N$), the force of attraction due to charges is $\sim 10^{-9}N$, but servations $\delta F \simeq mN$

Full Model

Model describes tip coordinates x and z (depth). Our equations are of the general form

$$m\ddot{y} = F_a - F_r \tag{7}$$

where y represents x or z,

 $F_a \longrightarrow$ applied force and $F_r \longrightarrow$ material response

- In equilibrium, $F = F_r$.
- In dynamic conditions, $F \neq F_r$ due to visco-elastic and plastic deformation.

Contributions to force :

- Projected area in x- and z-directions

$$A_x = A_{x,0} z^{3/2} \sim \frac{2}{3} \pi \sqrt{2R} z^{3/2}.$$

$$A_n = \pi a^2.$$

- Frictional force at the contactting interface is $A_{x,0}\tau_0 z^{3/2}$,
- au_0 is shear strength
- Frictional resistance arising from contact electrification = $\mu A_n \sigma^2/2\epsilon_0 \kappa$.
- Visco-elastic creep contribution to the flow = $\eta_{\parallel}A_z\dot{x}/D$: $A(z) = A_{x,0} z^{3/2}$.

Model

Plastic strain rate $\dot{\epsilon}_p = \dot{\epsilon}_0 (\tau/\tau_y)^n$ - Plastic deformation is continuous beyond the linear visco-elastic flow. $= \eta_{\parallel} A_{x,0} z^{3/2} \frac{\dot{x}}{D} \left[1 - \left(\frac{F}{A_{x,0} \tau_0 z^{3/2}} \right)^n \right]$

D Equation of motion along x – direction.

$$m\ddot{x} = F - F_r$$

$$m\ddot{x} = F - A_{x,0}\tau_0 z^{3/2} - \mu \pi R z \frac{\sigma^2}{2\kappa\epsilon_0}$$

$$m_x A(z) \frac{\dot{x}}{2} \left[1 - \left(\frac{F}{2\kappa\epsilon_0} \right)^n \right]$$
(8)

$$- \eta_{\parallel} A(z) \frac{x}{D} \Big[1 - \Big(\frac{1}{A_{x,0} \tau_0 z^{3/2} + \mu \pi R z \sigma^2 / 2\kappa \epsilon_0} \Big)^n \Big], \tag{9}$$

$$\dot{F} = K_{\parallel}(V_a - \dot{x}), \tag{10}$$

Model

The equation of motion along z- direction is

$$m\ddot{z} = F_n - \frac{4}{3}R^{1/2}z^{3/2} \Big[E^* + \frac{\eta_{\perp}}{D}\dot{z} \Big(1 - \Big(\frac{F_n}{\tau_{y,n}\pi Rz}\Big)^q \Big) \\ + \frac{c\eta_{\perp}}{D}\dot{x} \Big] - F\frac{z^{1/2}}{\sqrt{2R}}$$
(11)

The elastic response = $\frac{4}{3}R^{1/2}z^{3/2}E^*$.

 $\eta_{\perp} \dot{z}/D$ = visco-elastic creep along F_n .

Plastic strain rate = $-\frac{\eta_{\perp}}{D}\dot{z}\left(\frac{F_n}{\tau_{y,n}\pi Rz}\right)^q$. *q* exponent.

The tip creeps in the *z* direction, the position of tip center *x*, also creep along *x*. Additional creep contribution $c\eta_{\perp}\dot{x}/D$.

Note Eq. (8) gives $a^2 = [3RF_n/4E^*]^{2/3}$ Hertz radius.

Model

The evolution of the charge density σ is given by

$$\dot{\sigma} = \frac{\sigma_m}{t_a} \left(1 - \frac{\sigma}{\sigma_m} \right) - \frac{\dot{x}}{D} \sigma \tag{12}$$

 σ_m = maximum charge density.

 t_a = time constant.

Total charge $\sigma_t = \pi R z \sigma$,

$$\dot{\sigma}_t = \frac{(\pi R z \sigma_m - \sigma_t)}{t_a} + \sigma_t \frac{\dot{z}}{z} - \dot{x} \frac{\sigma_t}{D}$$
(13)

The Scaled Equations

 $D = F_{max}/K_{\parallel} \text{ and a time scale determined by } \omega^2 = K_{\parallel}/m.$

The scaled variables are : x = XD, z = ZD, $F^s = F/K_{\parallel}D$, $\bar{\tau}_0 = \tau_0 A_{x,0} D^{1/2}/K_{\parallel}$, $\bar{F}_n = F_n/K_{\parallel}D$, $K_{\perp} = \frac{4}{3}(RD)^{1/2}E^*$, $\bar{\eta}_{\perp} = \frac{4}{3}(RD)^{1/2}\omega\eta_{\perp}/K_{\parallel}$, $\bar{\eta}_{\parallel} = A_0\eta_{\parallel}\omega/K_{\parallel}D$, $\bar{\tau}_y = \tau_{y,n}\pi R/K_{\parallel}$, $v_a = V_a/\omega D$, and $T_a = t_a\omega$.

The Scaled Equations

$$\ddot{X} = F^{s} - \bar{\tau}_{0} Z^{3/2} - \mu \frac{\Sigma^{2}}{Z} - \bar{\eta}_{\parallel} \dot{X} \Big[1 - \Big(\frac{F^{s}}{\bar{\tau}_{0} Z^{3/2} + \mu \frac{\Sigma^{2}}{Z}} \Big)^{n} \Big],$$
(14)
$$\ddot{Z} = \bar{F}_{n} - \Big[\frac{K_{\perp}}{K_{\parallel}} + \bar{\eta}_{\perp} \dot{Z} \Big(1 - \Big(\frac{\bar{F}_{n} K_{\parallel}}{\bar{\tau}_{y} Z} \Big)^{q} \Big) + c \bar{\eta}_{\perp} \dot{X} \Big] Z^{3/2} - \beta F^{s} Z^{1/2},$$
(15)
$$\dot{\Sigma}_{m} Z - \Sigma - \dot{X} \Sigma + \dot{Z} \Sigma$$
(16)

$$\dot{\Sigma} = \frac{\Delta m \Sigma - \Delta}{T_a} - \dot{X}\Sigma + \frac{\Delta}{Z}\Sigma, \qquad (16)$$

$$\dot{F}^s = v_a - \dot{X}. \tag{17}$$

- The scaled equation for charge transferred (to the substrate) is $\dot{\Sigma}_d = \dot{X}\Sigma$.
- Stick-slip due to feedback loop.
- Steady state unstable Instead of stability analysis, calculate force in stationary condition

$$F^{s} = \frac{\bar{\tau}_{0}\bar{F}_{n}}{\left[\frac{K_{\perp}}{K_{\parallel}} + c\bar{\eta}_{\perp}v_{a}\right]}.$$
(18)

Parameters

R	m	K_{\parallel}	F_n	V_a	σ_m
(mm)	(kg)	(N/m)	(mN)	($\mu m/s$)	(C/m^2)
0.5	10^{-5}	47	68 - 106	~ 10	1.67×10^{-5}
E^*	η_{\parallel}	η_{\perp}	$ au_0$	$ au_{y,n}$	σ_0
GPa	Pa.s	Pa.s	MPa	MPa	(nC/m^2)
1 - 3		•••	0.1 - 10	1 - 50	3.0
K_{\perp}	$ar{\eta}_{\parallel}$	$ar{\eta}_\perp$	$ar{ au}_0$	$ar{ au}_y$	Σ_m
$\sim 10^6$	10^{2}	8×10^8	1.0	2500	0.0167

- The magnitude of $\tau_0 \sim \mu \tau_s$, where τ_s is the shear yield stress and $\mu \sim 0.3$ is the friction coefficient.
- Other parameters used are: $A_0 \sim a^2 \sim 10^{-10} m$, $\kappa = 3.3$, $v_a = 1.45 \times 10^{-5}$, $T_a = 2$ and c = 0.122. Choosing $F_{max} = 47 mN$ gives $D = 10^{-3} m$ and thus the range of \bar{F}_n corresponding to 68 - 106mN is 1.446 - 2.26.



- Correlation between stick-slip events and charge transfer.
- \blacksquare The ratio of maximum deviation (1.09 × 10⁴) to the mean (3.365 × 10⁴) of α is $\sim 30\%$.
 - Comparable with 25% maximum scatter for the range of $F_n = 68 106 \ mN$ in expt.



Thus, α^s independent of load is due to limited range of F_n , not due to true idependence. $A_n \propto F_n^{2/3}$, α depends on load.

 α^s same for smooth sliding, steady state.

Note $A_n \longrightarrow 1$ to 1.34, $F_n \longrightarrow 68$ to 106 mN. Not really independent but small ramge of F_n



For larger loads fewer events of larger magnitude (on an average) for the same scan length.

In steady state $\int K_{\parallel} \dot{x} dt = \alpha \int \dot{\sigma}_d dt = \alpha \int (\dot{x} \sigma_t / D) dt \longrightarrow K_{\parallel} D \simeq \alpha \pi R z_{asy} \sigma_{asy}$. Using z_{asy} and σ_m leads to $\alpha \sim 50 \ eV/\text{\AA}$ for $F_n = 94 \ mN$ which is even after discounting the ideal nature of the model.

- Possible reasons: Experimental charging area large; Roughness

- In reality in experiments, charging radius R_c is 10-20 times a for $\sigma \sim 1.6 - 2.4 \times 10^8 \ charges/mm^2$.

Thus, $A_c = \beta A_n$ with $\beta \sim (\frac{R_c}{a})^2 \sim 100 - 400$ or use higher charge density.

- Using βA_n in place of A_n , we get $\alpha \sim 0.5 - 0.125 \ eV/\text{\AA}$ that is consistent with the reported value.

- Roughness can also be contribute to typically 10%
- Static friction is less than sliding friction.

Summary and Conclusions

- The correlation between the stick-slip events and charge transfer reproduced.
- Lack of dependence of the scale factor α on normal load is due to the small range of values used in expt. Indeed, the scale factor is inversely proportional to the area of contact. Using $A_c = \pi a^2$ gives Using experimental contact area $A_{\varepsilon} = \beta A_c$, $\beta \sim 100 400$; give $\alpha = 0.25 0.5$ eV.
- These features follow naturally due to the separation of time scales of stick and slip events.
- As most experimental features are captured, the model provides an alternate explanation for the results.
- Stick-slip events in our model are not influenced by charge.
- Plastic deformation of the interfacial layer is responsible for stick-slip dynamics that arises due to a competition between the internal relaxation time scales (visco-elastic and plastic deformation time scales etc) and that due to the pull speed.

THANK YOU